

REMARKS

Claims 1-31 were pending in the application, with claims 30 and 31 being withdrawn from consideration. The amendment cancels claims 21-23 and adds new claims 32-38.

The Amendments to the Claims

Claim 1 has been amended to more clearly reflect what the Applicants regard as their invention in view of the variation in pore size measurements which can be produced by different measurement techniques. In particular, reference to the pore size of the support has been deleted. This amendment is supported at page 3, lines 8-9 and page 10, line 23 of the specification.

Claim 6 has been amended to specify that the pore size of the support is between about 0.2 micron and about 4 microns. These amendments are supported by original claim 1 and at page 16, lines 23-24. Claim 6 has also been amended to depend from claim 1 rather than claim 5.

Claim 19 has been amended to specify that the SAPO is SAPO-34. This amendment is supported by original claim 4. Claim 19 has been further amended to specify that the CO₂/CH₄ separation selectivity is greater than about 60 at a temperature of about 297 K for an approximately 50/50 CO₂/CH₄ mixture with about 222 kPa feed pressure and about 138 kPa pressure drop. This amendment is supported at page 14, lines 27-28 and at page 18, Table 1.

Claim 20 has been amended to specify that the SAPO is SAPO-34. This amendment is supported by original claim 23. Claim 20 has been further amended to delete reference to the pore size of the support. Since the claim refers to SAPO-34 crystals being present within the pores of the support, an indication of the pore size of the support is already present in the claim. Claim 20 has been further amended to specify that the CO₂/CH₄ separation selectivity

is greater than about 60 at a temperature of about 297 K for an approximately 50/50 CO₂/CH₄ mixture with about 222 kPa feed pressure and about 138 kPa pressure drop. This amendment is supported at page 14, lines 27-28 and at page 18, Table 1.

Please cancel claims 21-23 without prejudice.

Claim 25 has been amended to specify that the pore size of the support is between about 0.2 micron and about 4 microns. These amendments are supported by original claim 20 and at page 16, lines 23-24.

Claims 28 and 29 have been amended to delete reference to the specific SAPO, since the specific SAPO is now stated in claim 20, from which claims 27 and 28 depend.

New claim 32 depends from claim 1 and recites that the calcining temperature is between about 623 K and about 673 K. This claim is supported at page 12, lines 31-32 of the application.

New claim 33 depends from claim 1 and recites that the support is a porous metal support. This claim is supported at page 10, line 26 of the application.

New claim 34 depends from claim 5 and recites the limitation that the gel does not completely fill the pores of the support. This claim is supported at page 10, lines 17-19 of the specification.

New claim 35 recites a method for making a crystalline silicoaluminophosphate (SAPO) membrane, the method consisting essentially of the steps: a) providing a porous support; b) preparing an aqueous SAPO

forming gel comprising an organic templating agent; c) aging the gel; d) contacting the porous support with the aged gel; e) heating the porous support and the gel to form a layer of SAPO crystals on the surface of the support; f) washing and drying the support and the SAPO layer; and g) calcining the SAPO layer to remove the templating agent; wherein steps d) and e) are repeated until the SAPO layer is substantially impermeable to N₂ before performing step f) and when step d) is repeated the gel is contacted with the porous support and the previously formed layer of SAPO crystals. This claim is supported by original claims 1, 16, 18 and in the specification at page 3, lines 8-9, page 10, line 23, page 11, lines 26-30 and at page 12, lines 11-23.

New claim 36 recites a method for making a crystalline silicoaluminophosphate-34 (SAPO-34) membrane, the method comprising the steps of: a) providing a porous support having a pore size between about 0.2 microns and about 4 microns; b) preparing an aqueous SAPO-34 forming gel comprising an organic templating agent; c) aging the gel for at least 24 hours between about 290 K and about 300 K; d) contacting the porous support with the aged gel; e) heating the porous support and the gel to form a layer of SAPO-34 crystals on the surface of the support wherein the porous support and gel are heated to a temperature between about 465K and about 480 K; f) washing and drying the support and the SAPO-34 layer; and g) calcining the SAPO-34 layer to remove the templating agent, wherein the calcining temperature is between about 623 K and about 673 K; wherein steps d) and e) are repeated until the SAPO-34 layer is substantially impermeable to N₂ before performing step f) and when step d) is repeated the gel is contacted with the porous support and the previously formed layer of SAPO-34 crystals. This claim is supported by original claims 1, 9, 13, 16, 18 and in the specification at page 11, lines 26-30, page 12, lines 11-23, and at page 16, lines 23-24.

New claim 37 depends from claim 20 and recites that the support is a porous metal support. This claim is supported at page 10, line 26 of the application.

New claim 38 depends from claim 36 and recites that the support is a stainless steel support. This claim is supported at page 10, line 32 of the application.

It is believed that no new matter has been added by any amendment.

The Rejections Under 35 U.S.C. 102

Claims 1, 5-7, 9, 12, 13 and 18

Claims 1, 5-7, 9, 12, 13, and 18 were rejected under 35 U.S.C. 102(b) as being anticipated by Soria et al. (U.S. 6,472,016).

Soria et al. describes processes for preparing a membrane comprising a porous carrier and a layer of a molecular sieve. In these processes, a precursor solution, rather than a precursor gel, is employed. Since Soria et al. distinguish solution methods from gel methods at col. 1, lines 14-21, it is submitted that Soria et al.'s precursor solutions do not encompass precursor gels.

Since step b) of claim 1 of the present application involves preparing an aqueous SAPO forming gel, the Soria reference fails to teach all the limitations of claim 1. Applicants therefore respectfully request reconsideration and withdrawal of the rejection of claim 1.

Claims 5-7, 9, 12, 13 and 18 depend from and incorporate all the limitations of claim 1. Applicants further note that the Soria reference fails to teach the gel composition of claim 7. In view of all the foregoing, Applicants

respectfully request reconsideration and withdrawal of the rejection of claims 5-7, 9, 12, 13 and 18.

Claims 19, 20 and 24-27

Claims 19, 20 and 24-27 were rejected under 35 U.S.C. 102(b) as being anticipated by Soria et al. (U.S. 6,472,016).

Amended claim 19 relates to a SAPO-34 supported membrane made by the method of claim 1. Since Soria et al. do not specifically disclose SAPO-34 membranes, Soria et al. do not teach all the limitations of amended claim 19. Therefore, Applicants respectfully request reconsideration and withdrawal of the rejection of claim 19.

Amended claim 20 specifies that the membrane comprises SAPO-34 crystals. Since Soria et al. do not specifically disclose SAPO-34 membranes, Soria et al. do not teach all the limitations of amended claim 20. Therefore, Applicants respectfully request reconsideration and withdrawal of the rejection of claim 20. Since claims 24-27 depend from and incorporate all the limitations of claim 20, Applicants respectfully request reconsideration and withdrawal of the rejection of claims 24-27.

The 35 U.S.C. 103 Rejections

Claims 2-4, 8, 10 and 11

Claims 2-4, 8, 10 and 11 were rejected under 35 U.S.C. 103(a) as being unpatentable over Soria et al. (U.S. 6,472,016 B1) in view of Yoshikawa et al. (U.S. 6,503,294 B2). The Office Action states:

Soria et al. does not disclose the specific SAPO or the templating agent being tetraethyl ammonium hydroxide.

Yoshikawa et al. discloses a similar method of making a SAPO membrane wherein the SAPO comprises SAPO-34 and the templating agent is tetraethyl ammonium hydroxide in col. 24, line 48 to col. 25, line 4.

It would have been obvious to one of ordinary skill in the art at the time the invention was made to incorporate the SAPO-34 of Yoshikawa et al. into the method of Soria et al. to provide a membrane having a specific pore size for a desired separation, as is well known in the art. It would have been obvious to one of ordinary skill in the art at the time the invention was made to incorporate the TEOH of Yoshikawa et al. into the method of Soria et al. in that substituting one known templating agent for another would have been obvious to one having ordinary skill in the art.

At col. 24, line 48 through col. 25, line 4, Yoshikawa et al. describe a gel-based method for synthesizing a SAPO-34 membrane. Soria et al. relates to solution-based synthesis methods and teaches against the use of gel-based synthesis methods at columns 1 and 2. Therefore, there is no motivation to combine the gel-based methods of Yoshikawa et al. with the solution-based methods of Soria et al.

In addition, because the Soria and Yoshikawa references relate to dissimilar synthesis methods, simple substitution of Yoshikawa et al.'s synthesis gel into Soria's process may produce unpredictable crystallization results. In particular, in the methods of Soria et al. both an inert solution and a precursor solution of the molecular sieve are prepared and the porous carrier is completely impregnated with the inert solution and/or the precursor solution (Col.3, lines 40-43). As described, the inert solution is used to prevent deposition of molecular sieve on some regions of the carrier. The use of an inert solution as taught by Soria et al. in combination with Yoshikawa's gel would be expected to dilute Yoshikawa's gel composition, potentially producing unpredictable crystallization results. For example, combination of an inert solution with a precursor gel could change the zeolite crystal structure, nucleation rate, and/or growth rate from that expected from the precursor gel alone.

Furthermore, the combination of the Soria and Yoshikawa references do not provide motivation for aging Yoshikawa et al.'s synthesis gel prior to bringing

it in contact with the porous support. Yoshikawa et al. are silent with respect to aging of the synthesis gel. Soria et al. teach aging of the precursor solution in order to lead to the desired precursor solution (col. 9, lines 27-31), but do not provide specific teaching regarding aging of synthesis gels. Aging the gel is a limitation of claim 1, from which claims 2-4, 8, 10 and 11 depend.

In view of all the foregoing, Applicants respectfully submit that a prima facie case of obviousness has not been made out and request reconsideration and withdrawal of the rejection of claims 2-4, 8, 10 and 11.

Claims 14-17

Claims 14-17 were rejected under 35 U.S.C. 103(a) as being unpatentable over Soria et al. (U.S. 6,472,016) in view of the publication "Separation of Light Gas Mixtures Using SAPO-34 Membranes." The Office Action states:

Soria et al. does not disclose repeating the steps recited in claim 1. The publication "Separation of Light Gas Mixtures Using SAPO-34 Membranes" (hereinafter, "the publication") teaches repeating the steps until the membrane is impermeable to nitrogen at page 780, col. 2, lines 9-37.

The Poshusta publication "Separation of Light Gas Mixtures Using SAPO-34 Membranes" describes a gel-based method for synthesizing SAPO-34 membranes. As previously discussed, Soria et al. relates to solution-based methods for making zeolite membranes and teaches against the use of gel-based synthesis methods at columns 1 and 2. In particular, Soria et al. describe the need for supplementary deposition-crystallization cycles in gel processes as being a disadvantage (see col. 1, lines 32-37 and col. 1 line 66 through col. 2 line 2). Therefore, there is no motivation to combine the gel-based methods of the Poshusta publication with the solution-based methods of Soria et al.

In addition, because the Soria reference and the publication relate to dissimilar synthesis methods, simple substitution of the publication's synthesis gel into Soria's process may produce unpredictable crystallization results as discussed with respect to the Yoshikawa reference. Furthermore, the combination of the Soria reference and the publication does not provide motivation for aging the publication's synthesis gel prior to bringing it in contact with the porous support (for membrane formation). Therefore, Applicants respectfully request reconsideration and withdrawal of the rejection of claims 14-17.

Claims 21-23, 28 and 29

Claims 21-23, 28 and 29 were rejected under 35 U.S.C. 103(a) as being unpatentable over Soria et al. (U.S. 6,472,016) in view of Yoshikawa et al. (U.S. 6,503,294). The Office Action states:

With regard to claims 21-23, Soria et al. does not disclose the specific SAPO.

Yoshikawa et al. discloses a similar SAPO membrane wherein the SAPO comprises SAPO-34 in col. 24, line 48 to col. 25, line 4.

It would have been obvious to one of ordinary skill in the art at the time the invention was made to incorporate the SAPO-34 of Yoshikawa et al. into the membrane of Soria et al. to provide a membrane having a specific pore size for a desired separation, as is well known in the art.

With regard to claims 28 and 29, the combined membrane of Soria et al. and Yoshikawa et al. will inherently exhibit the recited selectivity and permeate concentration since the membrane has the same structure as the claimed membrane.

Applicants note that claim 20 has been amended to recite that the SAPO membrane is SAPO-34 and that claims 21-23 have been cancelled. Amended claim 20 also incorporates the limitation that the CO₂/CH₄ separation selectivity is greater than about 60 at a temperature of about 297 K for an approximately 50/50 CO₂/CH₄ mixture with about 222 kPa feed pressure and about 138 kPa

pressure drop. Claims 28 and 29 depend from and incorporate all the limitations of amended claim 20.

Applicants respectfully disagree that the suggested combined membrane of Soria et al. and Yoshikawa et al. will inherently exhibit the selectivity recited in claims 20 and 28 and the permeate concentration recited in claim 29. The selectivity of a crystalline zeolite-type membrane depends not only on the crystal structure of the zeolite but also on the presence of “nonzeolite pores” present in the membrane, as noted in the specification at page 15, lines 21-33. The Poshusta publication cited in the present Office Action shows that an increase in the number of non-SAPO-34 pores lowers the selectivity (page 787 of the publication). Therefore, the selectivity recited in claims 20 and 28 can be taken as an indication of the quantity of nonzeolite pores in the membrane.

Applicants respectfully submit that incorporation of the SAPO-34 of Yoshikawa et al. into the membrane of Soria et al. via combination of the processes of Yoshikawa et al. and Soria et al. will not necessarily result in formation of any SAPO-34 membrane on the porous support, much less a SAPO-34 membrane with the desired selectivity. As previously described, the use of an inert solution as taught by Soria et al. in combination with Yoshikawa's gel would be expected to dilute Yoshikawa's gel composition, potentially producing unpredictable crystallization results. Furthermore, there is no motivation to combine the gel-based methods of Yoshikawa et al. with the solution-based methods of Soria et al., since Soria et al. teach away from the use of gel-based methods.

In addition, the suggested combination of references is not enabling for SAPO-34 membranes of the desired level of CO₂/CH₄ selectivity since neither reference demonstrates this level of selectivity for SAPO-34 membranes. The Soria reference shows experimental results only for silicalite (MFI structure)

membranes; silicalite is known to the art to have a larger pore size than SAPO-34 (CHA structure). The Yoshikawa reference measures only a hydrogen selective permeability for the SAPO-34 membranes of col. 24, line 48 through col. 25, line 4. However, an estimation of the CO₂/CH₄ separation selectivity of the membrane of Yoshikawa et al. may be obtained through comparison with another Poshusta et al. publication currently of record in the application, "Synthesis and Permeation Properties of SAPO-34 Tubular Membranes". This 1998 Poshusta et al. publication uses a similar gel composition, the same crystallization temperature and a lower calcination temperature (753K vs. 773 K) than Yoshikawa et al. The 1998 Poshusta et al. publication indicates that the best room temperature CO₂/CH₄ separation selectivity obtained was 30, a factor of two less than is required by claim 20.

Applicants furthermore respectfully disagree that the suggested combined membrane of Soria et al. and Yoshikawa et al. will inherently exhibit the permeate concentration recited in claim 29. The permeate concentration will depend on the membrane CO₂/CH₄ separation selectivity. As discussed above, the combined membrane of Soria et al. and Yoshikawa et al. does not inherently have the required CO₂/CH₄ separation selectivity.

In view of all the foregoing, Applicants respectfully request reconsideration and withdrawal of the rejection of claims 28 and 29.

The New Claims

Claims 32-34 depend from and incorporate all the limitations of claim 1, which is believed to be in condition for allowance.

Claim 35 recites a method consisting essentially of the listed steps. Claim 35 is believed to be patentable over Soria et al. because Soria et al. does not disclose all the limitations of the claim. In particular, claim 35 refers to a

synthesis gel rather than a synthesis solution and does not include preparation or use of an inert solution as described by Soria et al. at Col.3, lines 40-41. Claim 35 is believed to be patentable over the combination of Soria et al. with Yoshikawa et al. or the Poshusta et al. publication because there is no motivation to combine solution-based synthesis methods with gel-based synthesis methods since Soria et al. teach away from the use of gel-based methods. Furthermore, the combination of Soria et al. with the other references does not provide motivation for aging a synthesis gel prior to bringing it in contact with the porous support.

Claim 36 is believed to be patentable over Soria et al. because Soria et al. does not disclose all the limitations of the claim. In particular, claim 36 refers to a synthesis gel rather than a synthesis solution. Claim 36 is believed to be patentable over the combination of Soria et al. with Yoshikawa et al. or the Poshusta et al. publication because there is no motivation to combine solution-based synthesis methods with gel-based synthesis methods since Soria et al. teach away from the use of gel-based methods. Furthermore, the combination of Soria et al. with the other references does not provide motivation for aging a synthesis gel prior to bringing it in contact with the porous support.

Claims 37 and 38 depend from and incorporate all the limitations of amended claim 20, which is believed to be in condition for allowance.

Conclusion

All claims being in condition for allowance, passage to issuance is respectfully requested. If there are further issues related to patentability, the courtesy of a telephone interview is requested, and the Examiner is invited to call to arrange a mutually convenient time.

It is believed that a fee of \$520 is due with this submission (including \$120 for a one month extension of time, \$200 for one independent claim in excess of three and \$200 for four additional dependent claims) It is intended that this fee will be paid electronically at the time of submission of this response. Please charge any deficiency or credit any overpayment of the fees accompanying this submission to deposit account 07-1969.

Respectfully submitted,
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